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Optimum Compositions for the Low-Temperature Fabrication of Highly Ordered FePt (001) and FePt (110) Films

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Abstract—The composition dependence of the degree of chemical order S and uniaxial magnetic anisotropy energy K_u has been investigated for FePt (001) and FePt (110) films deposited at the substrate temperature $T_s = 300^\circ\text{C}$, and the optimum compositional region for $L1_0$ ordering in FePt (110) films has been compared with that in FePt (001) films. It has been found that high S and large K_u are obtained around the stoichiometric composition for FePt (110) films, whereas they are obtained around an off-stoichiometric Pt-rich composition for FePt (001) films, indicating that the difference in optimum compositions for achieving highly ordered structure between FePt (001) and FePt (110) films at reduced temperature.

Index Terms—Composition dependence, FePt thin film, low-temperature fabrication, sputter-deposition.

I. INTRODUCTION

THE areal density of magnetic recording media has dramatically increased for the last decade [1]. In order to push the recording density up to the order of Tbit/in², the data have to be stored in magnetic particles of ~ 10 nm in diameter. However, the instability of magnetization due to thermal fluctuation is a serious problem for such nm-sized magnetic particles. One of the possible solutions for this problem is to use the magnetic materials with large magnetic anisotropy. In this context, $L1_0$ ordered FePt alloy with large uniaxial magnetic anisotropy ($K_u = 7.0 \times 10^7$ erg/cm³) [2] has attracted much attention as a candidate material to realize next-generation ultrahigh density magnetic recording media. Recently, a lot of work have been devoted to the fabrication of FePt thin films by sputter-deposition or molecular beam epitaxy technique [3]–[8]. A high temperature process such as deposition on a heated substrate and/or post-annealing above 500°C is usually required to form the $L1_0$ ordered structure for FePt films. However, such a high temperature process accelerates the grain growth of magnetic particles, leading to a low signal-to-noise ratio in recording media. Several attempts on the low temperature fabrication of FePt films were performed, such as the introduction of underlayers [9], the addition of third elements [10], [11], multilayering [12], [13], ion

irradiation [14], monatomic layer control [15], high Ar gas pressure during deposition [16], and *in situ* annealing [17]. In a previous paper [18], we reported that for FePt (001) films deposited on MgO (001) substrates high $L1_0$ order and high perpendicular magnetic anisotropy were successfully obtained by shifting the composition of the FePt phase to an off-stoichiometric Pt-rich region even in the case of the substrate temperature $T_s = 300^\circ\text{C}$, in contrast to the result around the stoichiometric composition showing poor chemical order.

In this paper, in order to investigate the effect of the crystal orientation on the optimum composition for the $L1_0$ ordering at reduced temperature, FePt (110) films with various compositions were prepared on MgO (110) substrates at $T_s = 300^\circ\text{C}$, and the optimum composition for high degree of order S and large K_u for FePt (110) films on MgO (110) substrates has been compared to that for FePt (001) films on MgO (001) substrates.

II. EXPERIMENTAL PROCEDURE

Films were prepared on MgO (110) substrates using a dc magnetron sputtering apparatus. Base pressure was below 1×10^{-9} torr, and high purity ($>99.9999\%$) argon of 5.0 mtorr was flown during sputtering. An Fe seed layer of 10 Å and subsequently a Pt buffer layer of 400 Å were deposited at room temperature. The FePt layer of 180 Å was prepared by co-deposition of Fe and Pt on the Pt buffer layer at $T_s = 300^\circ\text{C}$. The Fe concentration x (at.%) of $\text{Fe}_x\text{Pt}_{100-x}$ films was varied in the range from 19 to 68. The typical growth rate was 0.1 Å/s. The sample structure, compositions, and sputtering condition were the same as those reported in a previous paper [18]. The compositions of films were determined by electron probe X-ray microanalysis (EPMA). Structural characterization was performed by X-ray diffraction (XRD). Magnetic properties were measured by a superconducting quantum interference device (SQUID) magnetometer.

III. RESULTS AND DISCUSSION

XRD patterns for $\text{Fe}_x\text{Pt}_{100-x}$ films (grown on MgO (110) substrates) are shown in Fig. 1 [(a) $x = 68$, (b) 62, (c) 52, (d) 45, (e) 38, (f) 34, (g) 30, and (h) 19]. The fundamental 220 peaks of the FePt phase and the Pt buffer layer are clearly observed for all the films, indicating the films are strongly textured to the (110) plane. The unlabeled sharp peaks are due to the MgO substrate. For $x = 68$ and 62, only the FePt 110 superlattice peak with weak intensity is observed. With decreasing x to 52, the FePt 110 superlattice peak becomes intense and the FePt 330

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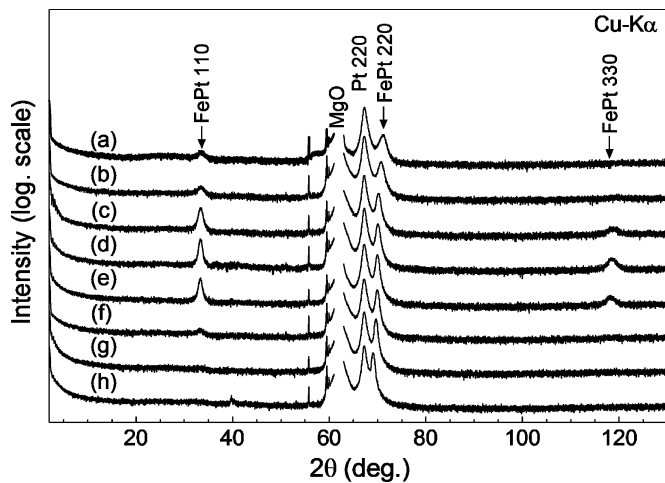


Fig. 1. X-ray diffraction patterns with Cu – K α radiation for FePt films grown on MgO (110) substrates at $T_s = 300$ °C. The Fe concentrations x (at. %) for FePt films are (a) 68, (b) 62, (c) 52, (d) 45, (e) 38, (f) 34, (g) 30, and (h) 19.

superlattice peak begins to appear. The sharp superlattice peaks indicating the formation of $L1_0$ ordered structure are observed at $x = 38$ –52. With further decrease of x , the superlattice peaks disappear. It is noted that $L1_0$ ordering of FePt (110) film is promoted around the stoichiometric composition at $T_s = 300$ °C, which is contrary to the results for FePt (001) films reported previously [18], i.e., the $L1_0$ ordered structure was best formed at an off-stoichiometric Pt-rich region for FePt (001) films. From the integrated intensities of fundamental and superlattice peaks extracted from numerical fitting, the degree of long-range chemical order S was evaluated. The detailed procedure for the evaluation of S was described in [4]. The maximum S of 0.7 ± 0.1 has been obtained for $x = 52$.

In order to confirm the epitaxial growth of FePt layer, two-dimensional XRD intensity mapping was performed. Fig. 2(a) shows the X-ray intensity mapping of {202} diffraction for the FePt (110) film with $x = 52$. For comparison, Fig. 2(b) shows the intensity mapping for the FePt (001) film with $x = 38$ [18], which is the optimum composition for the $L1_0$ ordering of FePt (001) films. The diffraction patterns were obtained from the FePt {202} planes illustrated as the gray areas. The scan directions of polar angle χ and rotation angle ϕ are also described in the inset of Fig. 2. For the FePt (001) film, the four-fold rotation symmetry around the [001] surface normal is clearly observed. On the other hand, two-fold rotation symmetry around the [110] surface normal is obtained for the FePt (110) film, reflecting the symmetry of the (110) plane of tetragonal $L1_0$ structure. These results indicate that the FePt (110) film is epitaxially grown on the MgO (110) single crystal substrate similarly to the FePt (001) film deposited on the MgO (001) substrate.

Fig. 3 shows the magnetization curves for FePt (110) films with $x =$ (a) 62, (b) 52, (c) 45, (d) 38, and (e) 34. Solid, broken, and dotted lines denote the magnetization curves measured along [001] and $[1\bar{1}0]$ directions in the film plane, and the perpendicular direction to the film plane, respectively. The directions of applied magnetic field are illustrated in the inset of Fig. 3. The crystal axes shown in the figures are those for the

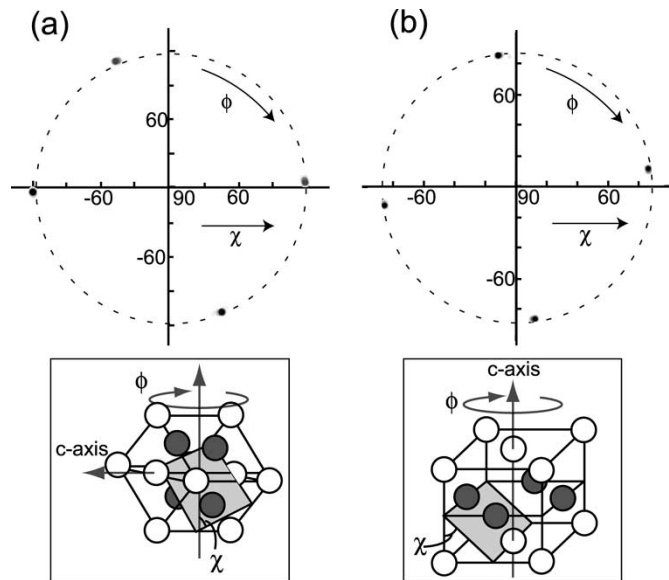


Fig. 2. X-ray intensity mappings for the {202} diffraction in (a) Fe₅₂Pt₄₈ (110) and (b) Fe₃₈Pt₆₂ (001) films. The FePt (202) planes are illustrated as the gray areas in the insets of the figures, and the scan directions of χ and ϕ are also denoted.

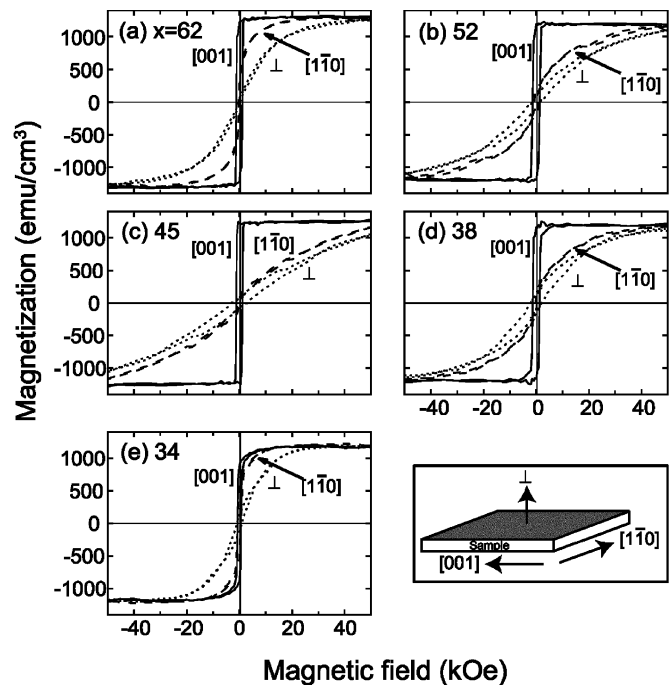


Fig. 3. Magnetization curves for FePt films with a variety of Fe concentration x (at. %) grown at $T_s = 300$ °C. x are (a) 62, (b) 52, (c) 45, (d) 38 and (e) 34. The magnetic field was applied along the [001] direction (solid line), the $[1\bar{1}0]$ direction (broken line) in the film plane, and the perpendicular direction to the film plane (\perp : dotted line) as illustrated in the inset.

MgO (110) substrate. FePt (110) films with large in-plane uniaxial magnetic anisotropy are obtained for $x = 38$ –52, and the easy magnetization axis lies along the in-plane [001] direction. The compositional region where large K_u is achieved coincides with that where superlattice peaks are clearly observed in Fig. 1. The uniaxial magnetic anisotropy energy K_u (with the [001] easy axis) was determined from the area enclosed

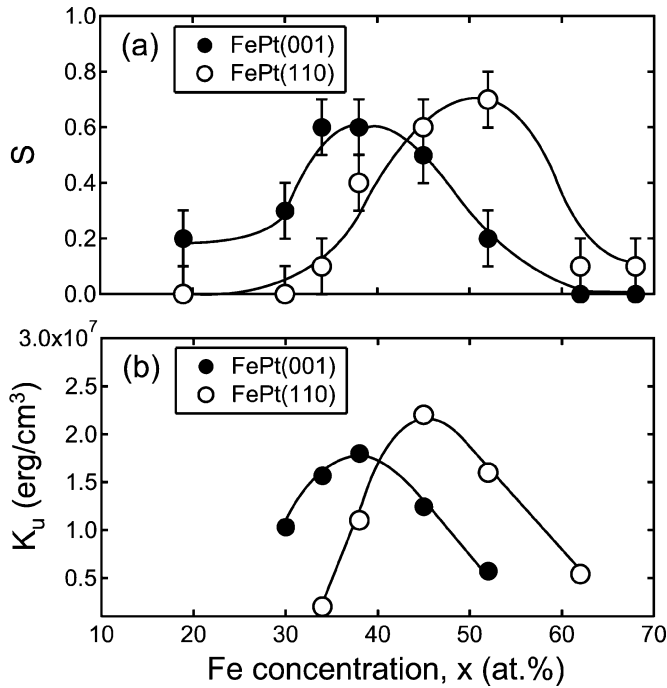


Fig. 4. (a) The degree of chemical order S and (b) the uniaxial magnetic anisotropy constant K_u for FePt films as a function of the Fe concentration x (at. %). Solid and open circles denote the results for FePt (001) and FePt (110) films, respectively.

between the magnetization curves with magnetic field applied along the [001] and $[1\bar{1}0]$ directions. The maximum value of $K_u = 2.2 \times 10^7$ erg/cm³ has been obtained for $x = 45$.

S and K_u as a function of x are summarized in Fig. 4(a) and (b), respectively. Open and solid circles denote the results of FePt (110) and FePt (001) films, respectively. For both the FePt (110) and FePt (001) films, the composition region for high S corresponds to that for large K_u . S and K_u hold maxima at an off-stoichiometric Pt-rich composition for FePt (001) films, whereas those hold maxima around the stoichiometric composition for the FePt (110) films. This indicates the difference in optimum compositions for high S and large K_u between FePt (001) and FePt (110) films. The origin for this difference is not clear at present, however, we may say that it is closely related to the lattice mismatch between the FePt layer and the Pt buffer layer. In the case of the FePt (001) film, the c-plane of the FePt layer is strained due to the mismatch with the Pt (001) buffer layer. On the other hand, the strained (110) plane for the FePt (110) film includes both the c-axis and the a-axis. The difference in the atomic planes that receive strain is considered to play an important role in the different optimum compositions for the $L1_0$ ordering between FePt (110) and FePt (001) films. It has also been found that the optimum compositional region for $L1_0$ ordering varies by using other under-layer materials for FePt (001) films, which will be reported elsewhere.

IV. CONCLUSION

$L1_0$ ordered FePt (110) films with large uniaxial magnetic anisotropy along the in-plane [001] direction were successfully prepared on MgO (110) substrates at $T_s = 300$ °C around the stoichiometric composition. The high S of 0.7 ± 0.1 and the

large K_u of 2.2×10^7 erg/cm³ have been obtained for $x = 52$ and 45, respectively, showing a clear contrast to the result for FePt (001) films where S and K_u showed maxima at an off-stoichiometric Pt-rich region ($x = 38$). In other words, the optimum composition for the $L1_0$ ordering in FePt films depends on the crystal orientation normal to the film plane.

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REFERENCES

- [1] D. Weller and A. Moser, "Thermal effect limits in ultrahigh-density magnetic recording," *IEEE Trans. Magn.*, vol. 35, pp. 4423–4439, Nov. 1999.
- [2] O. A. Ovanov, L. V. Solina, V. A. Demshina, and L. M. Magat, "Determination of the anisotropy constant and saturation magnetization, and magnetic properties of powders of an iron-platinum alloy," *Phys. Met. Metallogr.*, vol. 35, pp. 81–85, 1973.
- [3] B. M. Lairson, M. R. Visokay, R. Sinclair, and B. M. Clemens, "Epitaxial PtFe(001) thin films on MgO(001) with perpendicular magnetic anisotropy," *Appl. Phys. Lett.*, vol. 62, pp. 639–641, 1993.
- [4] A. Cebollada, D. Weller, J. Sticht, G. R. Harp, R. F. C. Farrow, R. F. Marks, R. Savoy, and J. C. Scott, "Enhanced magneto-optical Kerr effect in spontaneously ordered FePt alloys: Quantitative agreement between theory and experiment," *Phys. Rev. B*, vol. 50, pp. 3419–3422, 1994.
- [5] M. Watanabe and M. Homma, "Perpendicular magnetization of epitaxial FePt(001) thin films with high squareness and high coercive force," *Jpn. J. Appl. Phys.*, vol. 35, pp. L1264–L1267, 1996.
- [6] M. H. Hong, K. Hono, and M. Watanabe, "Microstructure of FePt/Pt magnetic thin films with high perpendicular coercivity," *J. Appl. Phys.*, vol. 84, pp. 4403–4409, 1998.
- [7] S. Okamoto, N. Kikuchi, O. Kitakami, T. Miyazaki, Y. Shimada, and K. Fukamichi, "Chemical-order-dependent magnetic anisotropy and exchange stiffness constant of FePt (001) epitaxial films," *Phys. Rev. B*, vol. 66, pp. 024413-1–024413-8, 2002.
- [8] T. Shima, K. Takanashi, Y. K. Takahashi, and K. Hono, "Preparation and magnetic properties of highly coercive FePt films," *Appl. Phys. Lett.*, vol. 81, pp. 1050–1052, 2002.
- [9] Y.-N. Hsu, S. Jeong, D. E. Laughlin, and D. N. Lambeth, "Effects of Ag underlayers on the microstructure and magnetic properties of epitaxial FePt thin films," *J. Appl. Phys.*, vol. 89, pp. 7068–7070, 2001.
- [10] T. Maeda, T. Kai, A. Kikitsu, T. Nagase, and J. Akiyama, "Reduction of ordering temperature of an FePt-ordered alloy by addition of Cu," *Appl. Phys. Lett.*, vol. 80, pp. 2147–2149, 2002.
- [11] Y. K. Takahashi, M. Ohnuma, and K. Hono, "Effect of Cu on the structure and magnetic properties of FePt sputtered films," *J. Magn. Mater.*, vol. 246, pp. 259–265, 2002.
- [12] C. P. Luo and D. J. Sellmyer, "Magnetic properties and structure of FePt thin films," *IEEE Trans. Magn.*, vol. 31, pp. 2764–2766, 1995.
- [13] Y. Endo, N. Kikuchi, O. Kitakami, and Y. Shimada, "Lowering of ordering temperature for fct Fe-Pt in Fe/Pt multilayers," *J. Appl. Phys.*, vol. 89, pp. 7065–7067, 2001.
- [14] D. Ravelosona, C. Chappert, V. Mathet, and H. Bernas, "Chemical order induced by ion irradiation in FePt (001) films," *Appl. Phys. Lett.*, vol. 87, pp. 236–238, 2000.
- [15] T. Shima, T. Moriguchi, S. Mitani, and K. Takanashi, "Low temperature fabrication of $L1_0$ ordered FePt alloy by alternate monatomic layer deposition," *Appl. Phys. Lett.*, vol. 80, pp. 288–290, 2002.
- [16] T. Suzuki, K. Harada, N. Honda, and K. Ouchi, "Preparation of ordered Fe-Pt thin films for perpendicular magnetic recording media," *J. Magn. Mater.*, vol. 193, pp. 85–88, 1999.
- [17] Y. K. Takahashi, M. Ohnuma, and K. Hono, "Low-temperature fabrication of high-coercivity $L1_0$ ordered FePt magnetic thin films by sputtering," *Jpn. J. Appl. Phys.*, vol. 40, pp. L1367–L1369, 2001.
- [18] T. Seki, T. Shima, K. Takanashi, Y. Takahashi, E. Matsubara, and K. Hono, " $L1_0$ ordering of off-stoichiometric FePt (001) thin films at reduced temperature," *Appl. Phys. Lett.*, vol. 82, pp. 2461–2463, 2003.